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Dynamic stormwater management to mitigate phosphorous export

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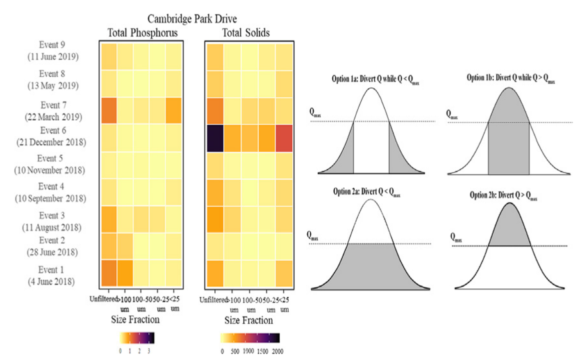
HIGHLIGHTS

- BMP based on TS as surrogate design parameter might not attain expected TP removal.
- TP & TS in stormwater are not uniformly distributed between particle size fractions.
- Diversion strategy targeting particles >25 μm have potential to remove >60%TP & >40% TS.
- Diverting half of the flow volume using flow diversion strategies removes 50% TP & TS.
- One feasible BMP design strategy is to combine both flow and particle size criteria.

GRAPHICAL ABSTRACT



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ABSTRACT

Altered stormwater flow characteristics and associated changes in nutrient and sediment fluxes due to urbanization threaten the water quality of many water bodies. For example, particle-bound phosphorus in stormwater runoff adds to the nutrient pool that can produce harmful algal blooms, and the associated particulate material can endanger fish and other living organisms in surface waters by increasing turbidity. While many studies have investigated how Total Solids (TS) particle size distributions vary in urban stormwater and the associated design criteria for Best Management Practices (BMPs) to remove TS, few studies have included different forms of phosphorus and their association with particle sizes to characterize design criteria to specifically maximize Total Phosphorus (TP) removal. This highlights a gap in our understanding of how the particle size distributions of TP and TS are related, and how these particle size distributions vary within and between storm events. Bridging this knowledge gap can improve design methods for BMPs specifically targeting phosphorus removal. This study characterizes within event (i.e., hourly) TP and TS particle size distributions and associated fluxes from urban catchments in the City of Cambridge, Massachusetts, to characterize potential TP and TS removal based on four different diversion and treatment strategies. The stormwater diversion strategies integrate new insights on temporal variations in particle size distributions and mass loading characteristics. In terms of diverted stormwater, the volume of stormwater treated tends to control more than what water is treated (e.g., first flush, event high flows, or smaller event flows). Based on the event data obtained in this study, considering flow volume different diversion approaches are optimal for TP vs TS, but treatment combined with particles sizes < 100 μm is best for both TP and TS.
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1. Introduction

Eutrophication, exacerbated by climate change and anthropogenic activity, is a significant problem impacting many surface water bodies (Kaushal et al., 2013; Oliver et al., 2019). Degraded water quality affects water treatment processes and recreational uses and is often linked to harmful algal blooms resulting from excess nutrients, such as nitrogen and phosphorus (Silva et al., 2019; Wang et al., 2019). Anthropogenic activities have increased the rate of phosphorus accumulation which is influencing eutrophication in many fresh water systems (Yan et al., 2016). In urban systems, particulate phosphorus accumulates on surfaces and is periodically transported to nearby rivers and lakes through stormwater runoff (Decina et al., 2018; Hobbie et al., 2017; Marsalek, 2014). Moreover, urbanization influences landscape modifications, affecting hydrologic and ecological dynamics (Jacobson, 2011; McGrane, 2016; Pickett et al., 2011). In general, urbanization increases impervious surface area that contributes to efficient transport of particulate and dissolved phosphorus (Janke et al., 2014; Kaushal and Belt, 2012; Petrone, 2010; Yu et al., 2012). The increase in phosphorus loading in the urban systems due to changes in landscape characteristics and use is comparable to the agricultural loading of phosphorus in rural systems (Duan et al., 2012; Omernik et al., 1981). To mitigate the impacts of urbanization and improve receiving water quality, a combination of structural and non-structural Best Management Practices (BMPs) are often used. In many cases, these BMPs (e.g. detention ponds, biofiltration system, sand filter basin, permeable pavements etc.) are designed based on gravimetric indices related to Total Suspended Solids (TSS) (Liu et al., 2017). However, our understanding of the particle size distributions characterizing event-based, urban stormwater runoff is limited, especially in terms of the phosphorus loading associated with discrete particle sizes.

While treating all urban stormwater runoff is not possible, one potential approach is to use flow control systems to selectively divert portions of stormwater with high phosphorus content to wastewater treatment plant or to smaller, in-situ, treatment systems. For example, a flow diversion device could be triggered by velocity, where threshold velocities are set based on shear stress values associated with movement of specific sizes of particulate material (i.e., based on total solids particle size distributions) in the stormwater pipes. However, adopting total solids as a surrogate for the design of these management strategies might not lead to the expected phosphorus removal (Obropta and Kardos, 2007). Particularly, in the case of flow diversion structures, where the design velocities are typically linked to TS particle sizes. To date, several studies have investigated forms of phosphorus and their associated particle size distributions (Kayhanian et al., 2012; Miguntanna et al., 2010; Shan et al., 2012; Zhang et al., 2018). For example, Zhang et al. (2018) found that the dominant forms of phosphorus (i.e., organic, inorganic, bio-available) in stormwater runoff vary with particle size. In an effort to understand total phosphorus wash-off potential, Vaze and Chiew performed wet filtration analysis on highway runoff and found that more than half of the TP was associated with particle sizes between 53 and 300 μm (Vaze and Chiew, 2004). However, there is a large variability in phosphorus transport and wash-off mechanisms based on the connectivity of impervious areas, land-cover, land-use and other biochemical characteristics (Beck et al., 2016). In addition, numerous studies have looked into total phosphorus loading within individual events to establish mass-limited pollutant runoff as a treatment facility design criteria (Memon et al., 2017; Yang and Toor, 2018; Zhang et al., 2013). However, to improve design methods for BMPs targeting phosphorus removal, we must address the knowledge gap in our understanding of how the particle size distributions of TP and TS are related, and how these particle size distributions vary within and between storm events. Therefore, the objectives of this study are to: (1) characterize the Particle Size Distributions (PSD) of Total Phosphorus (TP) and Total Solids (TS) in event stormwater runoff from select sampling sites distributed throughout the City of Cambridge, MA; and (2) assess the potential removal of pollutants from stormwater using dynamic flow diversion strategies. To support these objectives, hourly stormwater sampling was performed during nine storm events in four different watersheds with distinct land use characteristics over the period of June 2018 through June 2019. Note that, higher temporal frequency will provide more insight to the event pollutant variability. However, considering the constraint with sample bottle quantity and uncertainty associated with onset and duration of an event, to capture an entire event a uniform sampling protocol was maintained in the highly diverse study areas (ranging from 3.5 to 150 hectors). Again, the sampling approach presented in this study is best suited for discrete non-cohesive particles and intermediate-sized silts and neglects differences in density of solids with different particle sizes (high-density large particles like-sand, low-density organic particles from tree leaves, etc.).

2. Methods and materials

2.1. Study site

Urbanization and associated industrial, domestic and combined sewer discharges have significantly degraded water quality in the Lower Charles River, which flows through the Cities of Cambridge and Boston, MA, before terminating in Boston Harbor (Breault et al., 2002). Recently, efforts have been taken to eliminate these discharges and separate the existing combined sanitary and stormwater sewers to enable treatment of the sanitary flows. While the overall water quality in the Lower Charles River is improving, wet-weather water quality is still impaired due, in part, to pollutant wash-off (e.g., phosphorus) by the stormwater. For the Charles River, roughly half of the phosphorus in the river comes from industrial, commercial, and dense residential areas via stormwater runoff (Chapra et al., 2007). As part of the 1972 Clean Water Act, the National Pollutant Discharge Elimination System (NPDES) program was designed to help states reduce these discharges. This led to the development of the Massachusetts Municipal Separate Storm Sewer System (MS4) permit program. The MS4 permit requires that communities not exceed annual phosphorus loadings specified in any applicable Total Maximum Daily Load (TMDL) studies (Chapra et al., 2007). Specific to the Lower Charles River, this draft regulation requires the City of Cambridge, MA, to reduce its TP export by up to 62% of its estimated contributions during the period 1999–2000. To meet the TMDL allocations for phosphorous, the City developed and implemented a long-term mitigation plan. Currently, the City is installing structural and non-structural Best Management Practices to treat stormwater prior to entering any receiving waters.

In this study, storm water samples are collected from storm sewer (100–350 ft. upstream from the outfall) draining four urban areas with different land use characteristics (i.e., mostly residential and commercial) in the City of Cambridge, MA (Fig. 1a). These sites were selected based on their existing stormwater sampling infrastructure (i.e., discussed below). Among the sites, Cambridge Park Dr. and Columbus Ave. have a mix of residential and commercial development, while Western Ave. consists of mostly roadway drainage. The Cambridge Department of Public Works (CDPW) performs street sweeping once in a month from April to December throughout the city including the four study watersheds. The summary of drainage site characteristics is shown in Fig. 1b.

Data from two different rain gauges were used to estimate total event rainfall (Fig. 1). For Cambridge Park Dr. and Columbus Ave., the rain gauge near Fresh pond gate, Cambridge, MA (USGS Site ID: 422302071083801) was used. For Western Ave., the City of Cambridge's Department of Public Works rain gauge was used for the period June through December 2018, and the USGS Fresh pond gate, Cambridge, MA (USGS Site ID: 422302071083801) was used from Jan 2019 through June 2019. For this site, two rain gauges were used because the CDPW gauge was not operative during the winter season. All USGS stations

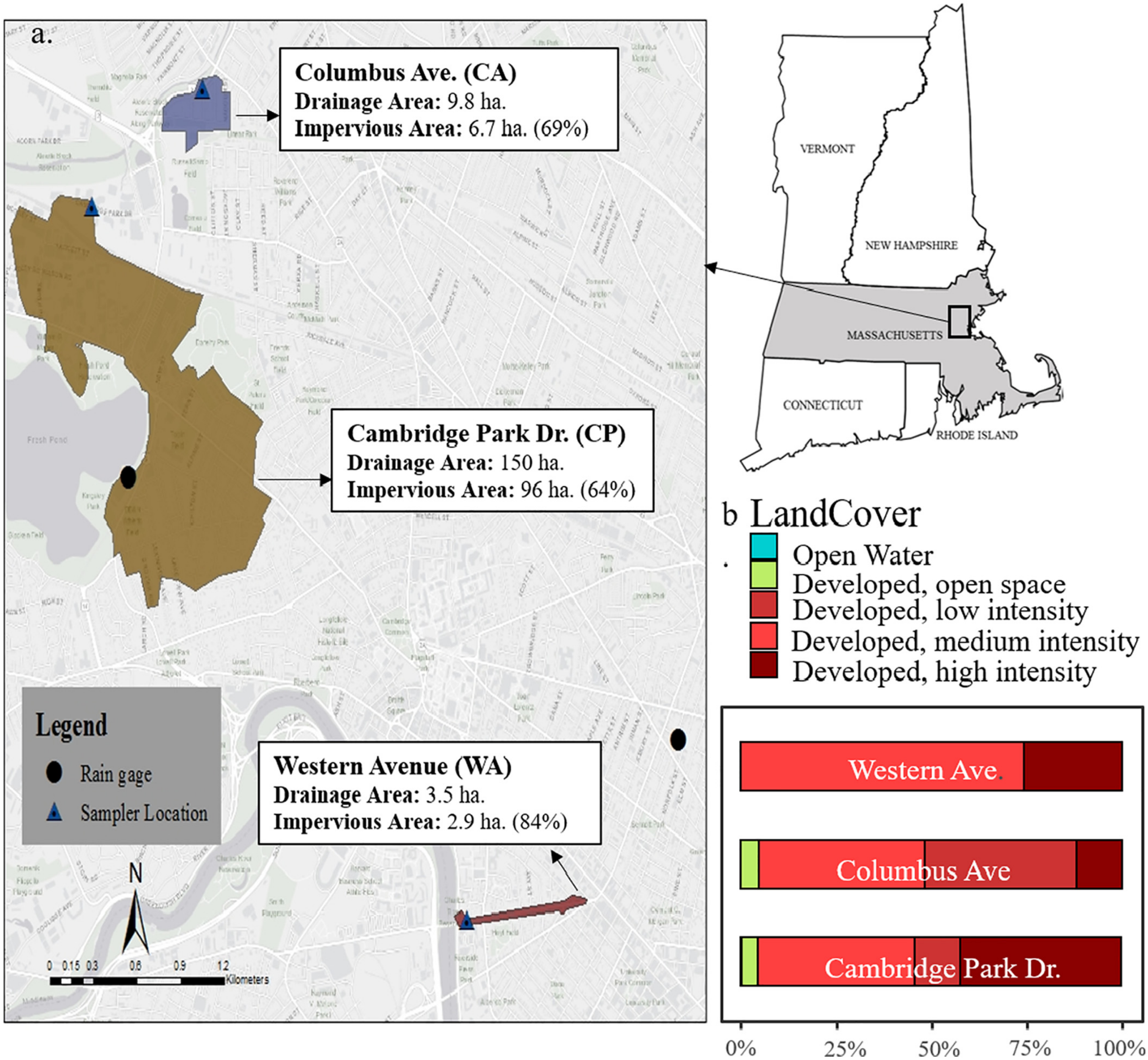

Fig. 1. (a) Study region showing four watersheds (shaded areas), two rain gauge and sampling locations in Cambridge, Massachusetts. (b) land cover distributions (MassGIS, 2016) for the study sites.

use standard rain gauge (model: Belfort 5–780) and Cambridge DPW uses tipping bucket rain gauge. Note that, differences in the underlying mechanism of different rain gauges and the distances between station locations might be responsible for different total rainfall depth for different watersheds in the same event.

2.2. Sample collection and analysis

The study watersheds, located within the City of Cambridge, MA, are primarily drained by a separate storm sewer system, with established monitoring locations just upstream of their outfall locations. The outfall for Western Ave tends to be partially submerged during dry weather due to the presence of a hydraulic control that maintains a water level in the outlet pipe but does not control during flow events, and all the other sites remain dry until stormwater runoff is generated during storm events. The sampling sites were located with the stormwater pipes roughly 100–350 ft. upstream from the outfalls, which minimizes the potential for backwater impacts. Each monitoring location consist of a permanent, weather-tight, above grade structure with a dedicated power supply containing all required monitoring equipment and instrumentation. Pumps and tubing are used to collect samples from the below grade stormwater drains. The lifting height of the samplers varied between 6 and 8 ft. (1.8–2.5 m) which is within the limit of lifting height for consistent effective sampling of particles less than 250 μm suggested by Clark et al., 2009. A combination of composite and non-composite event sampling was performed during the period June 2018 to June 2019. Note that, composite sampling was intended to maximize the duration of an event that could be sampled while collecting runoff at a relatively fine temporal resolution (e.g., 15-min). Composite sampling (1-hour or 45-minute composite samples based on 15-min, equal volume sub-sampling) was performed for six (6) storm events. While high temporal frequency (e.g., 5 to 15-min) or flow-weighted composite sampling is preferred, time-weighted composite sampling was implemented for the initial phase of the project due to uncertainty in storm durations and communications challenges with the autosamplers and flow measurement systems. After this initial period, to better understand the within event stormwater pollutant variability, sampling time-interval was reduced to 30 min and the sampling procedure was changed to discrete sampling for three (3) events. To assess the potential impacts on hourly flux estimates, the 30-min data from 3 events at CP and WA sites were used to compare hourly averaged and two consecutive 30-min TP fluxes. In general, the difference was less than 3%. The stormwater flows were sampled using Teledyne ISCO 6712FR Refrigerated Autosamplers using vinyl suction lines with a polypropylene strainer attached to the end to extract samples from the below grade storm sewers. A peristaltic pump draws the water sample through the strainers. Pre- and post-sampling purging ensures minimum cross-contamination between samples. Each autosampler was fitted with 24 1-liter polyethylene sample bottles. The sample bottles were acid washed before each sampling events. The triggering of the ISCO sampler was controlled using pre-set start date-time. The time to initiate the sampling procedure was determined based on the weather forecast prior to the event. As the weather forecasts get more accurate with an event approaching, the autosamplers were programmed 2–6 h before an event and the trigger was set accordingly. To avoid missing the starting of the event a few additional samples were collected even before the storm started. Irrespective of variable time of concentration at different sites, the auto samplers were manually triggered at the same time for all the sites for an event. Water samples were preserved in a 4 °C refrigerator immediately after collection. Note that, there were no baseflow observed in the study catchments. Additional event data such as flow, velocity and depth of flow were collected using a velocity meter at an interval of 15 min. Given the size difference between watersheds, 15-min flow data may not be sufficient to capture the full extent of variability in TS and TP fluxes between sites. Future sampling efforts will use 5-min flow rates.

Samples collected from June 2018 to December 2018 were divided into six subsamples based on particle size: unfiltered, filtered through 250, 100, 50, 25, and 10 μm filters. In 2019, samples were divided into unfiltered, filtered through 100, 50, and 25 μm and dissolved Phosphorus. Note that, the changes in filter sizes were based on the observed less variability in pollutant concentrations for particles filtered through 250 μm and 10 μm with the adjacent filter sizes (100 μm and 25 μm respectively). A total of 262 samples were collected resulting in 1458 subsamples being analyzed for TP and TS. The samples were then analyzed for TP using Hach kits, specifically kits for Total Phosphorus (Product # 2742645) using the PhosVer® 3 Ascorbic Acid method with Acid Persulfate Digestion: EPA Compliant Method 4500-P E (O'Dell, 1993).

To estimate the particulate solids load deflected to the WTP/similar treatment facilities from the flow diversion based urban stormwater treatment strategy, samples were also measured for TS according to APHA Method 2540- B: Total Solids Dried at 103–105 °C (APHA et al., 2005). Quality assurance and quality control (QA/QC) procedures included measurement of sample duplicates and reagent blanks. The relative percentage difference (RPD) ranged from 0 to 18% for TP. Method detection limit and target reporting limit for TP was 0.01 mg-P/L. Measured accuracy ranged from 80 to 120%. For TS every 10th sample was measured in duplicate. The RPD for TS were below 20%. Holding times for TP and TS measurement were 28 and 7 days, respectively. The results and analysis reported herein are based on the mean of each duplicate pair for TP and a combination of individual sample results and, when available, the mean of the duplicate pair for TS.

2.3. Land cover and land use

To characterize land cover and land use patterns, 1 m by 1 m resolution land cover data were obtained from the 2016 Land Cover Database (MassGIS, 2016). The data included categories for open water, developed open space, impervious area, forest, herbaceous/grasslands, emergent herbaceous wetlands, bare land etc. The City of Cambridge's high resolution photogrammetric Impervious Surface database was used to estimate the impervious areas within each drainage system (Cambridge GIS, 2010). These datasets were overlaid with the City of Cambridge's drainage area boundaries to extract site specific information (Fig. 1b).

2.4. Event mean concentrations and pollutant loads

To characterize the flow-weighted concentration of a pollutant for a given rainfall-runoff event, event mean concentration (EMC) is used. This is defined as total pollutant mass [M] discharged during an event divided by the total discharge volume (V) for the event (Sansalone et al., 1998), which is commonly used as an indicator parameter to evaluate and compare stormwater pollutants. The flow-weighted event mean concentration [M/V] is determined as:

$$EMC = \frac{M}{V} = \frac{\int Q(t)C(t)dt}{\int Q(t)dt} \qquad (1)$$

where C(t) is time variable pollutant concentration [M/V], Q(t) is time varying flow rate [V/T], and t [T] is the duration of the event.

Pollutant concentration expresses the strength of the pollutant, whereas pollutant loading considers the volume of water transporting the pollutant. This is a key parameter for BMP design as it provides an estimation of the volume that needs to be treated or diverted. Here, the numerator of Eq. (1) is defined as Pollutant Load, PL (kg).

2.5. Statistical analysis

To assess variations in TP and TS associated with different particle size fractions, the Kolmogorov-Smirnov test (K-S test) is performed for samples collected between June 2018 to December 2018 at 5% level of significance. Note that, samples collected in 2018 were divided into six subsamples based on particle sizes as described above. In 2019, dissolved Phosphate was included but the number of particle size fractions used to generate subsamples was reduced to four. Given only four size fractions for 2019 samples, the K-S test was only performed on six different particle size subsamples collected in 2018. Since water quality data do not follow conventional normal probability distributions, the non-parametric K-S test is used to compare the sample distribution with a reference probability distribution. The use of the K-S test for assessing the magnitude and variability TP and other water quality measures is common (Wang et al., 2017; Yu et al., 2016). For this analysis, a uniform distribution for TP and TS is assumed over all the particle size fractions, and the K-S test is used to assess this hypothesis. Here, a resulting *p*-value below the significance level (p-$value < 0.05$) indicates that pollutant load distribution in an event is not uniformly distributed between all particle size fractions. In other word, pollutant samples were not drawn from a uniform distribution. Note that, in case of small sample set for K-S test, the data points need to be highly different to reject the null hypothesis.

The Wilcoxon non-parametric test is performed to determine whether the differences in TP and TS concentrations between particle size fractions are statistically significant (p-$value < 0.05$). The temporally variable (30-min to 1-hour) concentrations associated with different particle size fractions for the sampled events and sites are used to assess differences between the particle size fractions. For different groups are used for this analysis: (i) events (i.e., time varying concentrations from each particle size fraction from all sites for a given event are grouped), (ii) sites (i.e., time varying concentrations from each particle size fraction from all events at a given site are grouped), (iii) combined events and sites (i.e., time varying concentrations from each particle size fraction from all events and all sites are grouped), and (iv) individual events and sites (i.e., time varying concentrations from each particle size fraction for a given event and site are grouped). The intent of using these four approaches is to assess variability in TP and TS concentrations associated with particle size fractions due to event, site and within event characteristics.

3. Results and discussion

3.1. Sampling events summary

Runoff samples for nine (9) storm events were collected between June 2018 and June 2019. The number of samples and the interval of sampling were based on weather forecasts. Table 1 provides a summary of the sampling events and rainfall characteristics.

3.2. Rainfall event analysis

In general, samples were collected only from events with total rainfall greater than roughly 13 mm. Given the event focus of this study, we used a threshold of 12.7 mm (0.5 in.) of rainfall to define events. A storm event smaller than this might be insufficient to generate enough runoff volume in the largest watershed (Cambridge Park Dr) to reach the sampling location. The peak intensity of these events ranged from 5.0 to 35 mm/h (Table 1). Event durations ranged from roughly 5.5 to 13 h. Antecedent dry periods, defined as the duration from the end of the last event that generated runoff to the beginning of the subject event, ranged from 1.5 to 19 days. Table 1 provides a summary of the rainfall characteristics for each event.

3.3. Variability in event TP and TS concentrations

Overall, TP and TS concentrations for unfiltered samples varied from 0.02 to 6.84 mg-P/L and 26 to 5110 mg/L, respectively, from all events and sites. Although WA has the smallest drainage area, its TP and TS concentrations are typically the largest (maximum unfiltered concentrations of TP and TS of 6.84 mg-P/L and 1060 mg/L, respectively: Tables S-2 and S-3), possibly due to its high imperviousness and short travel distances to the storm drain. Note that, WA was not sampled for event 6, which was the highest TS event at both CP and CA, but in general, WA TS concentrations are similar or higher than the other sites. The stormwater from this site primarily drains a nearby roadway. Dirt, biogenic particles (leaves, grass clippings) and roadway surface pollutants get washed into the storm sewer during storm events (Duan et al., 2014; Yang and Toor, 2017). In addition, automobile traffic characteristics are often correlated with total solids concentrations in stormwater runoff (Winston and Hunt, 2017). The particulate fraction of TP is also high at WA with only about 21% of TP represented as dissolved Phosphorus. In terms of seasonality, in this study, the largest TP and TS concentrations occurred in Dec-Mar period: max TP from WA (6.84 mg-P/L) in Mar 2019 (Event 7) and max TS from CP (5110 mg/L) in Dec 2018 (Event 6), which is highlighted in Fig. 2. Note that, the study area experiences an annual average snowfall of 48 in. in the period December through March. While it is a common practice to use gritting materials (e.g., sand) to treat roadways during winter storms in parts of Massachusetts, the City of Cambridge does not use gritting materials. Previous studies have tied the application of de-icing chemicals and the accumulation of pollutants in the snowpack during the winter period to an increase in stormwater pollutant load (Valtanen et al., 2014). A study in Minnesota showed that decomposition of accumulated leaf litter during late fall/winter contributes almost half of the annual total dissolved Phosphorus loading (Bratt et al., 2017). The situation gets worst during rain-on-snow events, where flushing of soluble and particulate Phosphorus from the impervious surfaces and snowpack increases the overall pollutant load. Similar to the study performed by Helmreich et al. (2010), seasonal influence on solids concentrations is observed from these sites. These results suggest there is an opportunity for pollutant reduction methods to focus on mitigating the impacts of late winter/early spring runoff events (e.g., increased street sweeping as soon as snow is not interfering) when resources are available.

Fig. 2 shows heatmaps of EMC for TP and TS separated by particle size. The average EMC for unfiltered TP at CA (0.29 mg-P/L) is nearly equal to the average EMC reported in Nationwide Urban Runoff Program (NURP) which is 0.33 mg-P/L (Pitt et al., 2004). However, at the other three sites, EMC is 1.5–3.0 times larger than reported values (e.g., WA-0.91 mg/L, CP–0.44 mg/L and SF-0.60 mg/L). The results for EMC-TP show that site and event characteristics influence the concentration magnitudes and variability between particle sizes (Table S-2). For example, event 7 results in the largest EMC-TP being associated with particles >100 μm at WA as compared to <25 μm at CP. The variability in particle size distributions (PSD) between sites may be explained by differences in hydrodynamics characteristics, such as time of concentration (Berretta and Sansalone, 2011). For example, WA has a shorter travel distance and smaller time of concentration (30-min)

Table 1
Summary of the sampled events.

Site	Event	Date	No. samples	Duration (h)	Total rainfall (mm)	Peak rainfall intensity (mm/h)	Antecedent dry days (days)	EMC-TP (mg/L)	EMC-TS (mg/L)
Cambridge Park Dr.	1	6/4/2018	10	8.5	12.5	5.1	14.5	0.93	428
	2	6/28/2018	8	7.5	29.2	34.5	3.18	0.48	110
	3	8/11/2018	8	5.5	26.9	29.5	1.48	0.61	492
	4	9/10/2018	13	11	24.6	9.14	19.1	0.19	407
	5	11/9/2018	10	7.3	27.4	11.2	2.94	0.10	182
	6	12/21/2018	15	10.25	16.5	7.11	4.29	0.18	2200
	7	3/22/2019	24	11	14.2	6.60	6.21	1.02	660
	8	5/13/2019	21	11	15.5	5.59	0.88	0.17	259
	9	6/11/2019	18	8.5	17	11.7	13	0.31	267
Western Ave.	1[a]	6/4/2018	6	7.67	17.5	6.86	14.5	1.97	682
	2[a]	6/28/2018	6	7.33	29	33.5	3.5	0.51	98.3
	3[a]	8/11/2018	7	4.67	16	16.0	1.48	0.22	326
	4[a]	9/10/2018	13	13.3	34.8	12.2	19.1	0.33	258
	5[a]	11/9/2018	9	8.00	38.1	12.2	2.94	0.19	119
	7	3/22/2019	16	11	14.2	6.60	6.21	2.82	1060
	8	5/13/2019	14	11	15.5	5.59	0.88	0.30	340
Columbus Ave.	1	6/4/2018	10	8.5	12.5	5.1	14.5	0.62	479
	2	6/28/2018	8	7.5	29.2	34.5	3.18	0.20	33.6
	5	11/9/2018	10	7.3	27.4	11.2	2.94	0.10	31.3
	6	12/21/2018	15	10.25	16.5	7.11	4.29	0.06	1540
	7	3/22/2019	21	11	14.2	6.60	6.21	0.48	71.5

USGS Site ID: 422302071083801, Fresh pond gate, Cambridge, MA rain gauge.
[a] Cambridge Department of Public Works rain gauge.

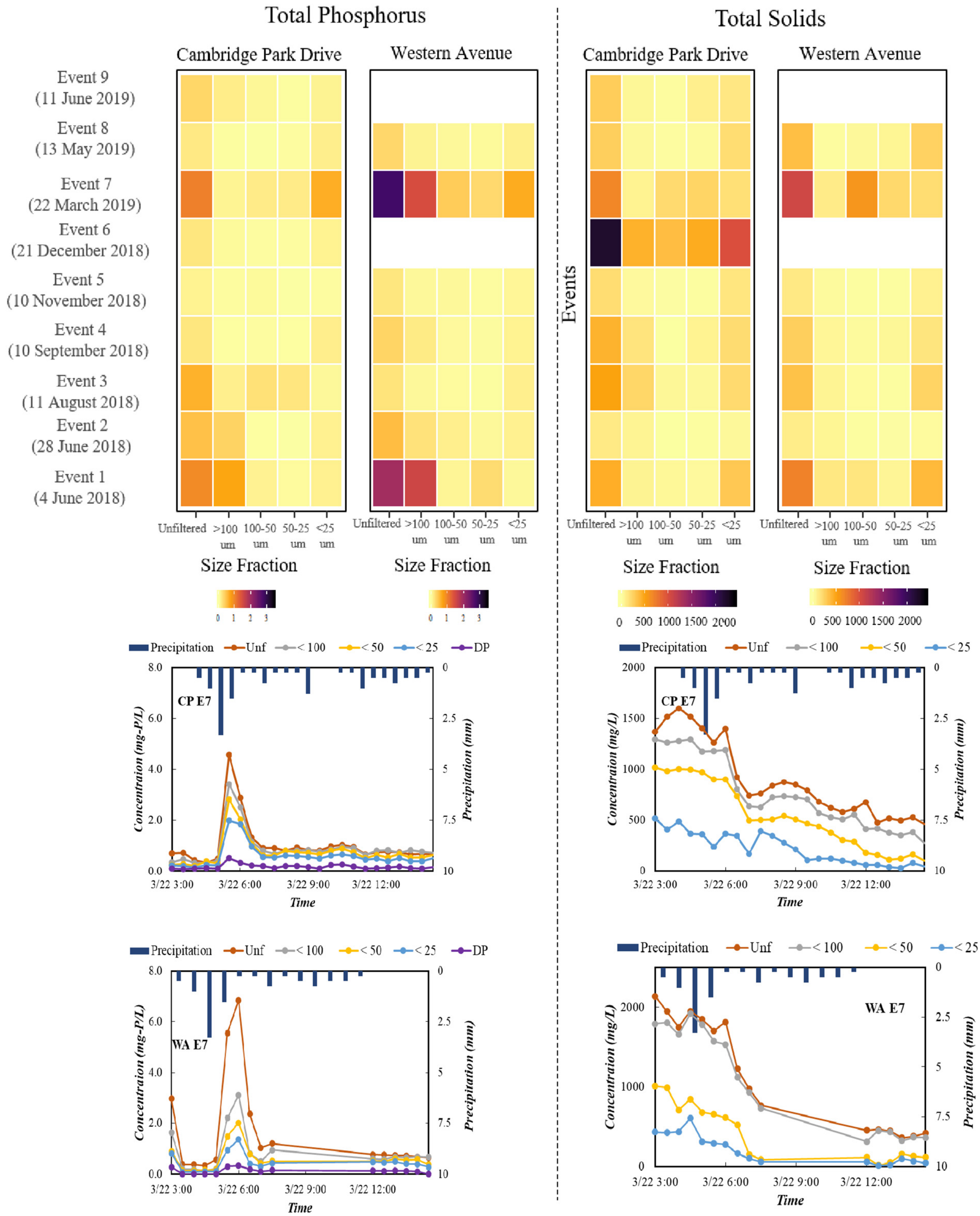

Fig. 2. (top) Heatmap of TP and TS event mean concentrations at CP and WA; (bottom) time varying TP and TS concentrations associated with different particle size fractions at CP and WA with rainfall (mm) for event No. 7 (March 2019).

to reach the sample collection point as compared to CP (180-min). Thus, larger biogenic (i.e., typically >75 μm, Berretta and Sansalone, 2011) particles associated with TP from WA are less impacted when arriving at the sampling location (i.e., closer to their original size). Whereas, for CP, these large particles travel a further distance and are more likely to be abraded. Moreover, the CP catchment has large, relatively low gradient drainpipes that tend to accumulate sediment, which might also be a factor in the size variation. In contrast to TP, EMC-TS is associated with particle size <25 μm at all the sites as highlighted in Fig. 2. This is consistent with a previous study by Zhao et al. (2010) that found 70% of the suspended solids from road deposited sediments were associated with finer particles. The regular street sweeping activity performed in the City of Cambridge might be the reason for the larger fraction of smaller sized particles on the streets (Amato et al., 2010). Overall, the EMC for TS varied from 31 to 2200 mg/L from all events and sites (Table S-2).

3.4. Correlation between EMC and event rainfall characteristics

The relationships between EMCs for different particle sizes (TP & TS at CP and WA) and event characteristics were assessed using a Pearson correlation analysis (Figure-S-1). Event characteristics included total rainfall (TR), hourly peak rainfall intensity (PRI) from 15-minute interval rainfall data, event duration (DUR), and antecedent dry days (ADD) (Table 1). Based on the correlation matrix analysis, CP EMC-TP and EMC-TS do not show strong correlation with event characteristics. Rather, the watershed size and complex land use and land cover pattern likely affects the variability in EMCs. Therefore, direct impact of the rainfall characteristics on the EMC for CP cannot be determined. However, WA EMC-TP for particle size 50–100 μm was strongly correlated with PRI ($r = 0.89$). A larger peak rainfall intensity led to an increased TP concentration associated with 50–100 μm particles that can be explained by the increased intensity producing runoff rates required to move larger particles. The strong positive correlation was also found between EMC-TSs of all sizes (except <25 μm) and ADD ($r_{>100\mu m} = 0.69$, $r_{100\text{-}50\mu m} = 0.57$, $r_{50\text{-}25\mu m} = 0.99$) suggesting the importance of particulate matter build up over time (Table S-3). The presence of smaller particles in all events irrespective of the dry period duration explains the weak correlation between EMC-TS of particle sizes <25 μm and ADD ($r = 0.02$).

3.5. Characterization of pollutants associated with different particle sizes

To understand the dynamics of TP and TS partitioning, the Kolmogorov-Smirnov test was performed using the TP and TS loading associated with six different particle size fractions. Eight of seventeen (~50%) event TP loading distributions showed statistically significant non-uniformity ($p\text{-value} < 0.05$) between different particle size fractions. In contrast to TP, six of seventeen (35%) event TS loading distributions showed statistically significant non-uniformity ($p\text{-value} < 0.05$) between different particle size fractions. The result of this analysis on high temporal resolution stormwater samples suggests that TP is not uniformly distributed between the particle size fractions, which supports potential stormwater diversion and treatment approaches based on flow velocity.

In addition, the Wilcoxon non-parametric test was used and shows that TP and TS concentrations for the various particle sizes are significantly different from each other (Table S-4). The average TP concentration at CP is dominated by the particle size fraction less than 25 μm. However, the largest mean TP concentrations at the other three sites are associated with particle sizes >100 μm. In contrast, the largest TS concentrations are ubiquitous for all the sites and associated with particles <25 μm.

Due to the high imperviousness of many urban areas, pollutant flux or load is a key component considered in BMP design, especially when the pollutant of interest is responsible for chronic effects (e.g. Phosphorus) rather than causing acute effects (e.g. fecal coliform or other toxicants). Here, the distribution of the total pollutant load (TP or TS) is illustrated in Fig. 3. Irrespective of landcover and site imperviousness, the TS load is predominantly associated with particle sizes <25 μm with a greater variability in the TP load distributions. A key finding highlighted in Fig. 3 is that the percentages of TP and TS associated with a given particle site fraction are not the same. This is especially important to note in the context of BMP designs based on particle sizes. If TP removal efficiency is intended, the BMP cannot be designed based on TS particle sizes. Designing a BMP to remove a specific particle size based on TS characteristics, can still result in significant TP loading due to smaller sized particles (Sample et al., 2012).

3.6. Potential removal efficiency based on fractional flow volumes

Previous studies have shown that a reasonable TP removal efficiency can be achieved using a flow deflection mechanism (Bedoya et al., 2014). This approach is enabled with a flow-regulating valve designed to divert flows above a specified value. Based on this concept, we assess four potential flow diversion strategies as shown in Fig. 4. The first strategy will divert all flows at the start and end of the storm (Fig. 4, Option-1a) by closing the flow-diversion valve while the flow exceeds a set maximum flow rate (Q_{max}). The second strategy will divert all flow volume in the middle of the event (Fig. 4, Option-1b) by opening the flow-diversion valve while the flow exceeds Q_{max}. The third and fourth strategy are a combination of weir and valve to divert flows above or below a set maximum (Fig. 4, Option-2a and Option-2b). Note that, a minimum flow threshold (Q_{min}) is used to initiate the diversion process (i.e., indicate a storm is occurring) for all strategies. Although not part of this study, a fully dynamic approach that switches between strategies during an event based on real-time flux measurements of phosphorous would be ideal.

To implement strategies in Fig. 4, site/event specific Q_{max} ranges from 2 to 20 $m^3/s/km^2$ (Table S-5) and were derived based on the assumption that 50% of the event flow volume can be diverted. In general, diverting 50% of the event flow volume results in roughly 50% TP and 50% TS removal efficiencies for all four strategies (Fig. 5). Note that, the assumed diversion of 50% of the event flow volume is arbitrary, the net reduction in TP is similar to the ultimate TP reductions needed for the City of Cambridge to meet their TMDL targets. However, the diversion strategies can be scaled up/down if treatment capacities for receiving diverted stormwater are known.

While the median TP and TS removal efficiencies were roughly 50% for all four strategies (Fig. 5), options 1a&b generally had a larger range in removal efficiency, especially for TP, as compared to options 2 a & b. This variability is likely due to the variability in pollutant loading and the duration of time for which the diversions occur. For example, options 1 a &b, which divert all flows for a given portion of the event, result in the diversions occurring for less time during the event as compared to option 2. However, option 1b had the most varied removal efficiency with standard deviation ranged from 6%–20% at different sites.

3.7. Potential removal efficiency based on particle size

With increasingly more stringent phosphorus load reduction regulations, many municipalities are looking for alternative treatment strategies. For example, as a part of their non-structural treatment strategy, the City of Cambridge maintains a monthly street sweeping schedule from April to December. The Cambridge Department of Public Works (CDPW) performs street sweeping once in a month throughout the city. However, given the relatively short durations between events, the frequency may need to be increased to obtain noticeable reductions. Plus, Massachusetts Plant Nutrient Regulation 2012 specifies the restrictions on the non-agricultural use of Phosphorus containing fertilizers during winter (Dec 1 to Mar 1). However, due to poor infiltration capacity of the blue-clay soil in the City Cambridge, BMPs related to improving infiltration are not suitable for the area (Skehan, 2001). One alternative solution is to use a passive control flow deflection device to divert stormwater flows to treatment facilities. To ensure removal for the targeted particle size, the associated fluid shear stress at the deflection point should be attained/exceeded to prevent settling of the particles. A typical BMP design assumption based on settling mechanism considers total suspended solids (TSS) as a proxy parameter (Ferreira and Stenstrom, 2013). This assumption is applied to determine flow conditions for which a particle of a specific size (suspended particles) will start moving within the pipe. As stated in Bedoya et al. (2014), the flow deflection mechanism could be triggered by the pipe flow velocity based on the targeted shear stress required to suspend solid particles of a specific size. According to that study particles below 45 μm have been washed off when the fluid shear stress is below a certain threshold. Therefore, triggering the flow diversion system after this threshold stress will take consideration of discrete non-

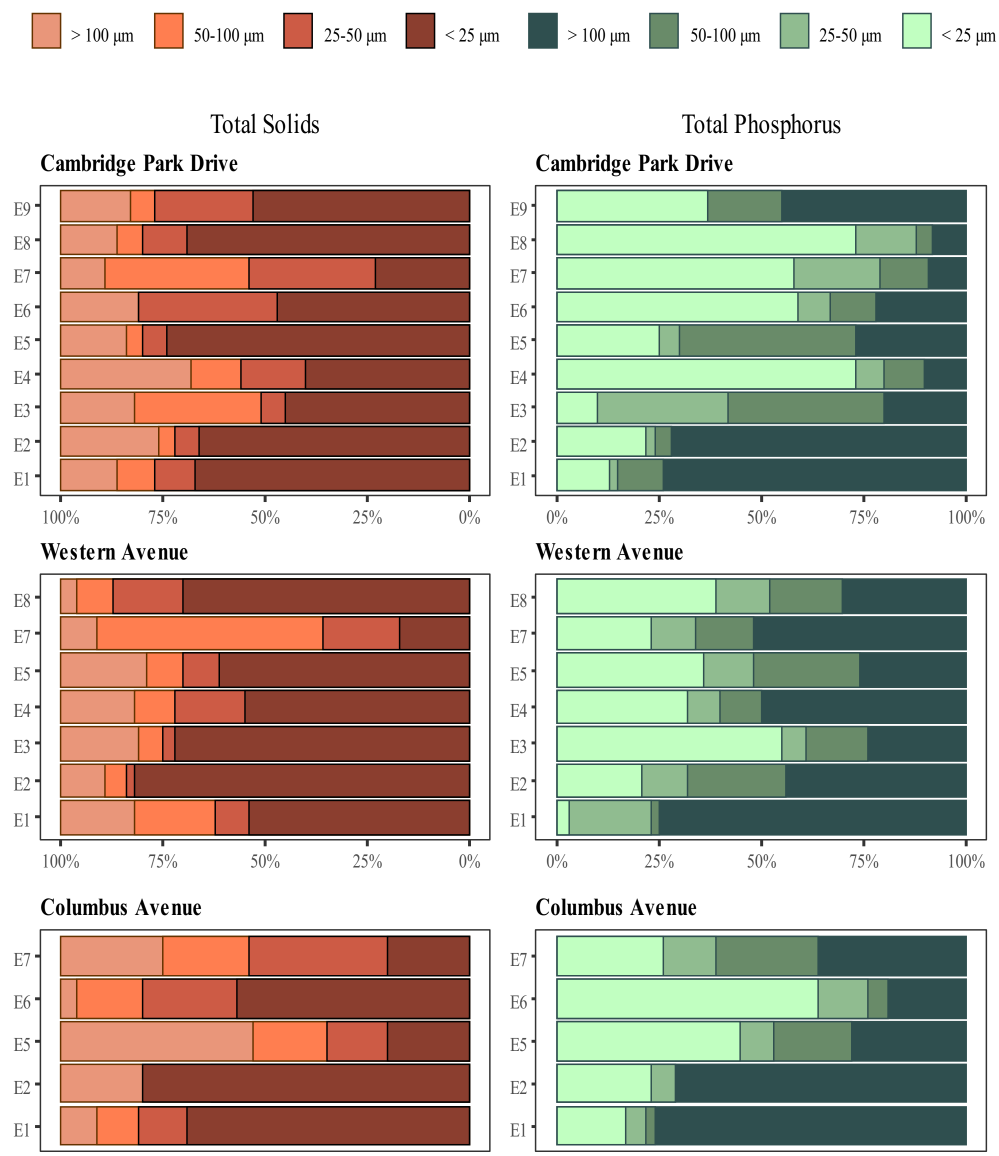

Fig. 3. Pollutant export distributions (%) based on particle size fractions at CP, WA and CA.

cohesive particles. However, in reality, solids removal efficiency of BMPs fail to reach the expected theoretical efficiency based on this mechanism since particle size distributions are not considered during design (Ferreira and Stenstrom, 2013). Similarly, while designing structural BMPs (e.g., stormwater detention basins), not taking TP particulate size distribution into consideration directly affects the eventual TP removal efficiency. A BMP designed to remove one particle-size fraction will likely fail to capture the potentially significant TP loads associated with the other size fractions (Fig. 6).

Building on this limitation, we investigated how pollutant export varies by particle size throughout events (i.e., cumulative pollutant export vs. cumulative flow) to identify individual particle size fractions containing the most TP and TS. In general, there is a significant difference in the percentage of event TP and TS export for a given particle size fraction (Fig. 6). Thus, the particle size fraction targeted in a BMPs design will lead to different levels of removal efficiency for TP and TS. For example, particles >100 μm account for 22–44% of the TP export but <25% of the TS export. Based on the sites/events sampled in this study, a treatment strategy targeting particle sizes >25 μm could attain a 63–74% TP and a 39–47% TS removal efficiency. The key findings are that the TP and TS particle size distributions are different, and the TP particle size distributions, not the TS distribution, should be used when designing phosphorous removal BMPs targeting a particular particle size(s).

3.8. Potential removal efficiency based on particle size and event flow fractions

Building on the above, we investigate both pollutant loads based on particle size factions and associated event flow volume fractions. This combined approach is especially important because capturing and diverting all event stormwater is not feasible for typical treatment facilities, which are generally already subjected to higher flow loadings during wet weather events. For example, the diversion approaches shown in Fig. 4 can be implemented based on target particles sizes (i.e., flow velocities/discharges for specific shear stresses), event volumes and/or specific fractions of event volumes. The diverted stormwater can then be sent to a BMP designed to remove particles larger than a specific particle size (i.e., based on settling time) or potentially a water treatment facility with near complete removal of all particles.

For the potential combined approach, Fig. 7 highlights how pollutant export varies with particle size thresholds (i.e., > or <100 μm, 50 μm and 25 μm) and each event flow volume quartile. In general, the

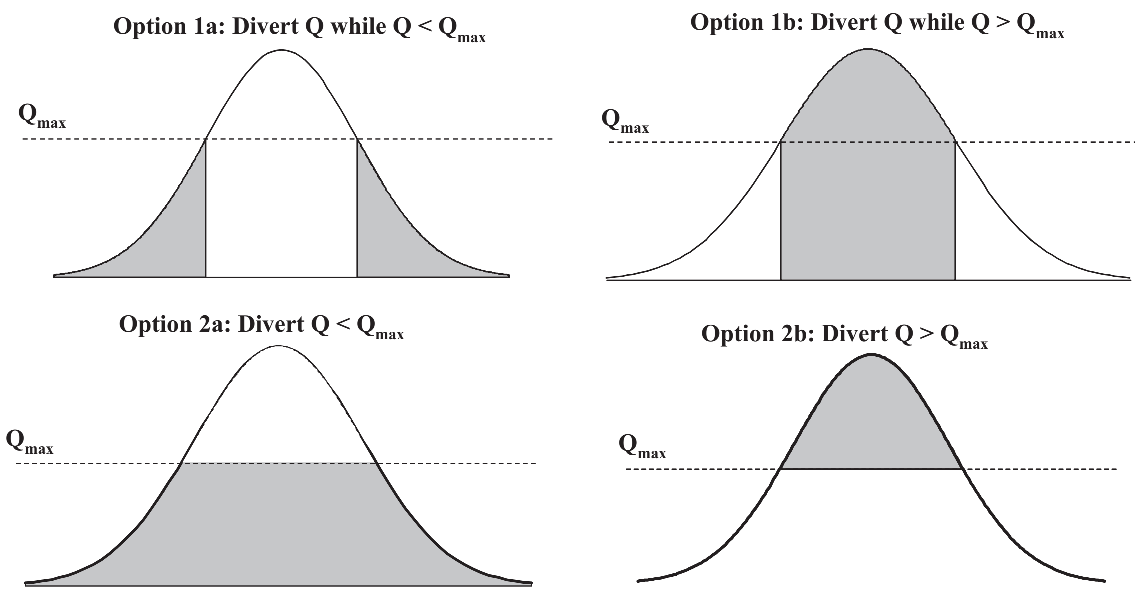

Fig. 4. Potential removal strategies based on flow rates (flow for the shaded area will be diverted).

maximum TP and TS loads are transported by the first 25% of the flow. A gradual decrease in pollutant export begins after the 1st flow quartile. The additional increase in TP flux in the second quartile (25–50%) of the flow at WA (88% impervious area) suggests a large amount of the load is coming from locations in the more upstream portions of the drainage basin. In the case of stormwater diversion or treatment structure designs constrained based on runoff volume capacity,

Fig. 7 provides a basis for determining which fraction of event runoff exports the maximum pollutant loads. This criterion is especially useful in cases where the watersheds do not clearly exhibit a first-flush phenomenon. Due to the complexity of pollutant wash-off criterion and dense urban land use/land cover patterns, a mass first-flush of pollutants is not a common observation (Morgan et al., 2017). Rather, combining BMPs designed to remove specific size fractions with diversion systems targeting specific event runoff volume fractions has the potential to improve TP removal efficiencies. For example, removing particle sizes >25 μm from the first 25% of the event flow would have removed 16–20% TP and 12–18% TS considering the sites in this study.

Characterization of nutrient flux associated with particle size fractions can provide water quality managers with insights on the shear stresses necessary for suspending and transporting particles of a particular size. When combined with the understanding of pollutant loads

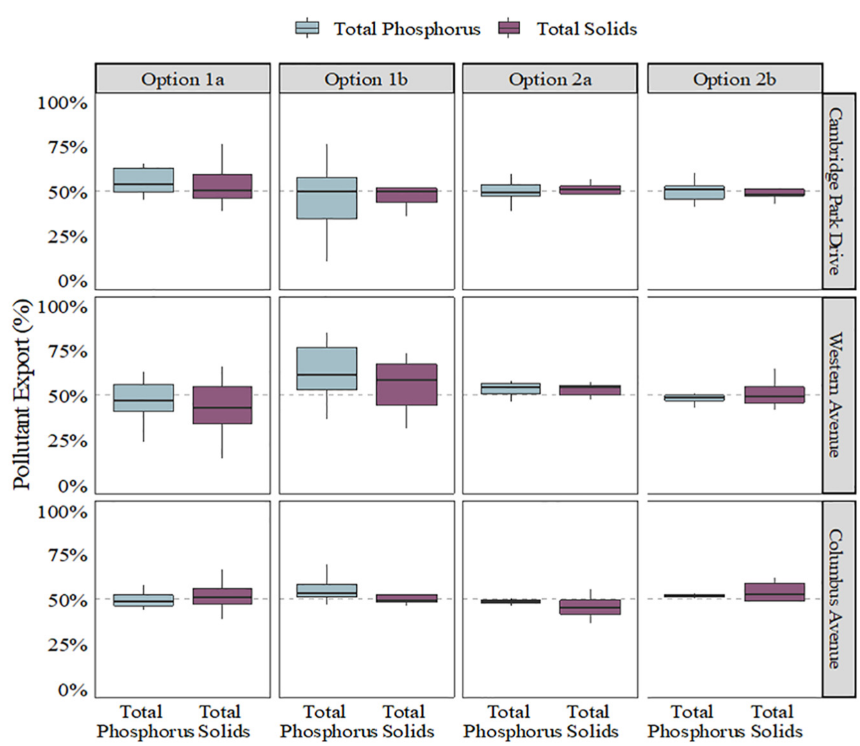

Fig. 5. Pollutant removal efficiency by the potential flow-based removal strategies.

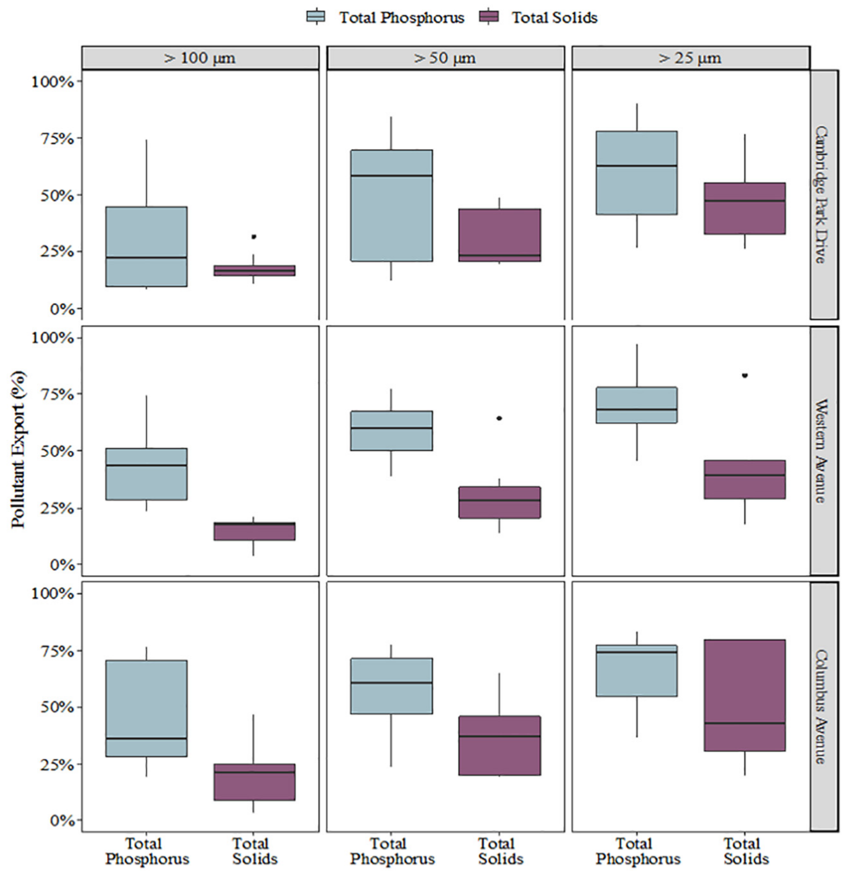

Fig. 6. Fraction of pollutant (TP or TS) loading based on particle size factions.

distributed throughout a runoff event, potential flow diversion BMPs can be optimized to maximize pollutant removal while minimizing the required treatment volume. Building on both flow and particle size results, the four diversion strategies discussed above are implemented conceptually and evaluated for potential TP and TS removal. Note, here we assume BMPs can be designed to remove participles greater than or less than a specified size. Thus, the four diversion scenarios represent eight cases, where particles > or < a specified size are removed from the diverted stormwater (Table S-6). The best options for TP removal are 1a or 2a < 100 μm and 1b or 2b > 25 μm. For example, using option 1a or 2a to divert 50% of the stormwater and remove particle sizes <100 μm reduces TP by 34% on average. Similarly, using options 1b or 2b to divert 50% of the stormwater and remove particles >25 μm results in roughly a 32–35% reduction in TP. For TS, diversion options 1b and 2b targeting removal of particles less than <100 μm results in the highest TS reductions (44%). However, there is a large reduction in TS removal when considering any option that removes particles greater than a given size class (i.e., a large fraction of TS is associated with particles <25 μm).

Note that, in above analysis, we assume that the hypothetical BMPs can remove TP and TS for a given particle size fractions by 100%. To expand the application of this study to other systems, the presented results can be scaled for actual BMP treatment efficiencies (e.g., 70–80% particles >75 μm) to estimate BMP specific treatment potential. It is also important to note that, in general, TP removal is best when targeting particles > 25 μm or <100 μm. However, removing particles less than specific size fractions (e.g., 50 or100 μm) tends to be best for TS removal Overall, treating particles <100 μm is the best for both TS and TP removal, which can be challenging for BMPs typically designed to remove particles greater than a specific size fraction (i.e., detention basins designed using settling velocities). In addition, while the results are somewhat variable, optimal diversion strategies (e.g., diverting fist flush, high flows, smaller event flows) tend to vary by site and differ for TP and TS. In general, when considering flow volume, the TP and TS reduction from the different diversion strategies are similar (i.e., the volume of water treated is more important than what portion of the event response is treated), but when combined with treatment of targeted particle size fractions, optimal, consistent removal of TP or TS can be obtained (i.e., target particles <100 μm).

4. Conclusions

For many communities, large reductions in phosphorus export are needed to improve surface water quality requiring novel best management practice approaches. One possible solution is developing stormwater diversion BMPs targeting select particle sizes and/or event flow fractions. Although it is not feasible to divert all event flows to treatment facilities, strategies can be developed to maximize pollutant removal while minimizing treated stormwater volume, where the upper limits on diverted stormwater are based on treatment facility capacities. For example, in this study, to reduce total phosphorus export by 65%, which is roughly the long-term target reduction for the City of Cambridge, MA, half (55%) of the event stormwater volume would need to be diverted to a treatment plant. To refine the overall fraction of event stormwater diverted for treatment, future research is needed to understand the causes behind the observed large variability in particle size fractions having the largest event mean phosphorus concentrations (i.e., varied from >100 μm to <25 μm between sites/events). This understanding is especially important given that a similar variability was not observed for total solids, which was consistently dominated by particle sizes <25 μm. Given the need for phosphorus export reductions by many communities, the results from this study highlight the need for additional research characterizing phosphorus particle size distributions in event stormwater and the development new BMPs that

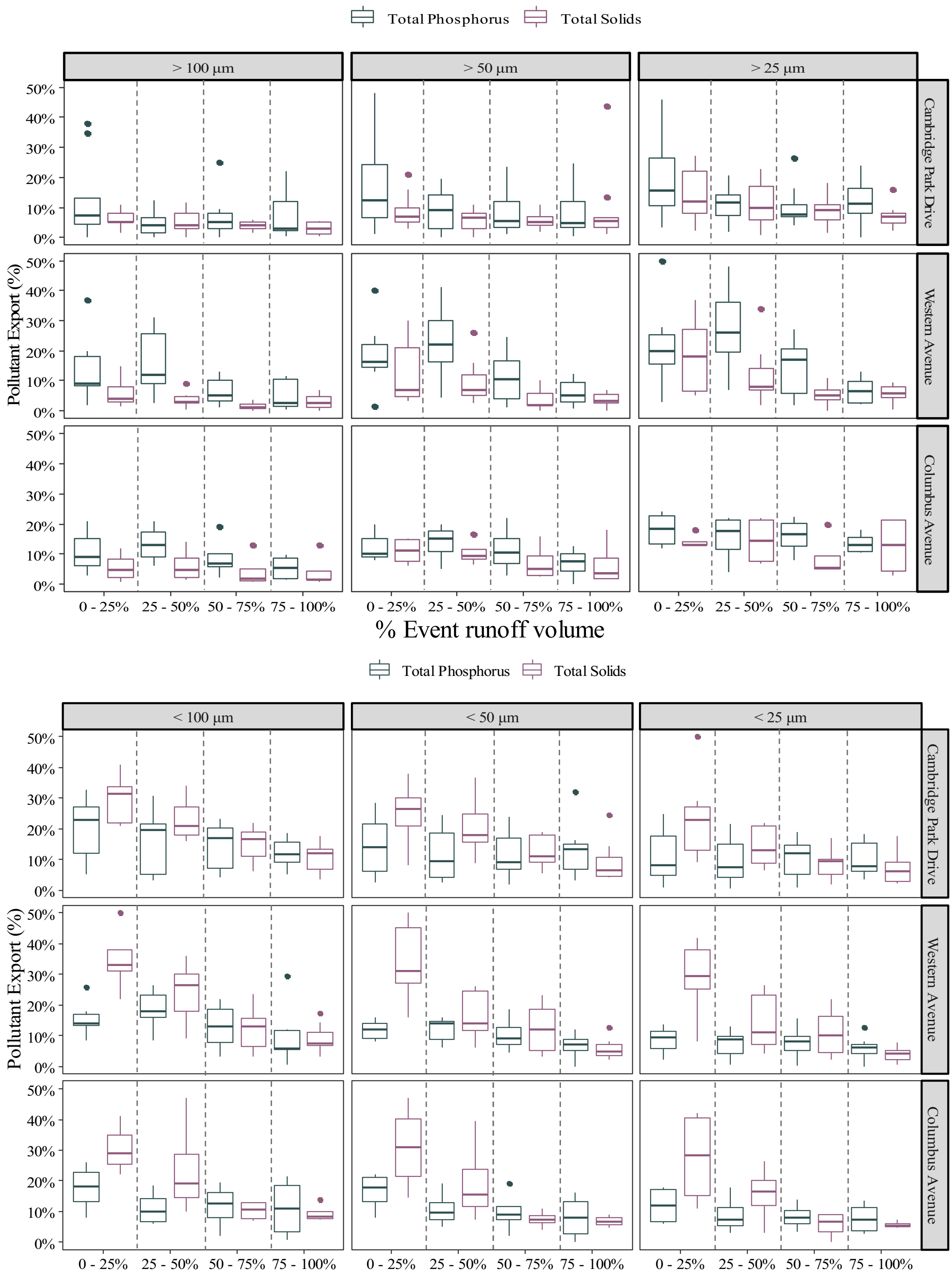

Fig. 7. Variability in Total Phosphorus (TP) and Total Solids (TS) export associated with particle sizes: (top) greater than 100 μm, 50 μm and 25 μm; or (bottom) less than 100 μm, 50 μm and 25 μm.

can leverage the differences in phosphorus and solids particle size distributions. While the results varied by event and site, the highlighting a management approach that targets flow and particle size fractions for optimal TP and TS removal. This approach can be expanded upon using real-time sensors for truly dynamic diversion and treatment.

CRediT authorship contribution statement

Sadia Tamanna Khan: Formal analysis, Investigation, Writing – original draft, Writing – review & editing, Visualization, Methodology. **R. Edward Beighley:** Supervision, Conceptualization, Methodology, Writing – review & editing. **David VanHoven:** Supervision, Project administration, Funding acquisition. **Kathy Watkins:** Supervision, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.147506.

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